



TITLE:

# <International Research Center for Elements Science>Photonic Elements Science

AUTHOR(S):

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CITATION:

<International Research Center for Elements Science>Photonic Elements Science. ICR Annual Report 2015, 22: 56-57

ISSUE DATE:

2015

URL:

<http://hdl.handle.net/2433/209853>

RIGHT:

# International Research Center for Elements Science – Photonic Elements Science –

<http://www.scl.kyoto-u.ac.jp/~opt-nano/index-e.html>



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## Scope of Research

Our research interest is to understand optical and quantum properties of nanometer-structured materials and to establish opto-nanoscience for creation of innovative functional materials. Space- and time-resolved laser spectroscopy is used to study optical properties of semiconductor quantum nanostructures and strongly correlated electron systems in low-dimensional materials. The main subjects are as follows: 1) investigation of optical properties of single nanostructures through the development of a high-resolution optical microscope, 2) development of nanoparticle assemblies with new optical functionalities, and 3) ultrafast optical spectroscopy of excited states of semiconductor nanostructures.

### KEYWORDS

Femtosecond Laser Spectroscopy  
Carbon Nanotubes  
Semiconductor Nanoparticles  
Transition Metal Oxides  
Semiconductor Nanostructures

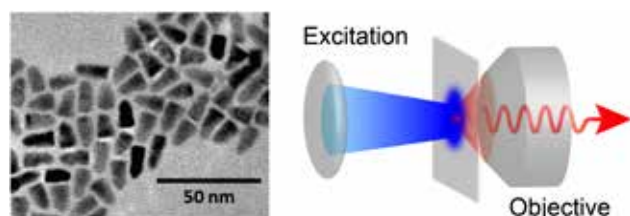


## Selected Publications

Ihara, T.; Kanemitsu, Y., Absorption Cross-section Spectrum of Single CdSe/ZnS NanoCrystals Revealed through Photoluminescence Excitation Spectroscopy, *Phys. Rev. B*, **92**, [155311-1]-[155311-5] (2015).  
Yamada, Y.; Yamada, T.; Le, P. Q.; Maruyama, N.; Nishimura, H.; Wakamiya, A.; Murata, Y.; Kanemitsu, Y., Dynamic Optical Properties of  $\text{CH}_3\text{NH}_3\text{PbI}_3$  Single Crystals as Revealed by One- and Two-photon Excited Photoluminescence Measurements, *J. Am. Chem. Soc.*, **137**, 10456-10459 (2015).  
Yamada, Y.; Nakamura, T.; Endo, M.; Wakamiya, A.; Kanemitsu, Y., Photocarrier Recombination Dynamics in Perovskite  $\text{CH}_3\text{NH}_3\text{PbI}_3$  for Solar Cell Applications, *J. Am. Chem. Soc.*, **136**, 11610-11613 (2014).  
Yamada, Y.; Sato, H. K.; Hikita, Y.; Hwang, H. Y.; Kanemitsu, Y., Measurement of the Femtosecond Optical Absorption of  $\text{LaAlO}_3/\text{SrTiO}_3$  Heterostructures: Evidence for an Extremely Slow Electron Relaxation at the Interface, *Phys. Rev. Lett.*, **111**, [047403-1]-[047403-5] (2013).  
Matsunaga, R.; Matsuda, K.; Kanemitsu, Y., Observation of Charged Excitons in Hole-doped Carbon Nanotubes Using Photoluminescence and Absorption Spectroscopy, *Phys. Rev. Lett.*, **106**, [037404-1]-[037404-4] (2011).

## Excitation Spectroscopy of Photoluminescence Properties of Single CdSe/ZnS Nanocrystals

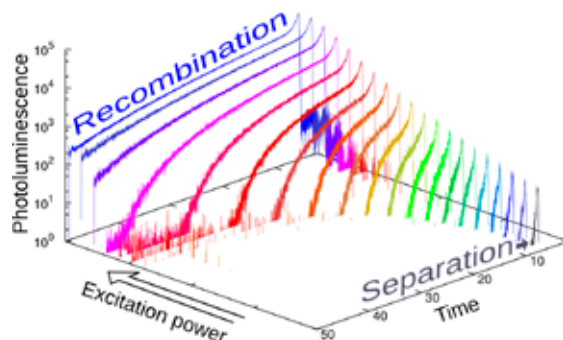
The absorption spectrum represents an important aspect of single semiconductor nanocrystals (NCs), revealing their morphology and orientation. By conducting quantitative photoluminescence excitation spectroscopy based on a single-photon counting method, we succeed in clarifying the absorption cross-section spectrum of neutral excitons in single CdSe/ZnS NCs. With the absorption cross section, we reveal that the intensity saturation of the neutral-exciton emission can be explained by the Poisson statistics for incident photons and by the small quantum yield of the biexciton emission. We also quantitatively evaluated the biexciton emission contribution and the ionization rate of NCs that appear at increased incident photon fluxes.



**Figure 1.** A TEM image of the sample and a schematic diagram of the excitation geometry.

## Contactless Characterization of Electrical Subcell Performance for Tandem Solar Cells

Tandem solar cells absorb different spectral parts in several subcells, which are grown in a multilayer structure. World-record conversion efficiencies have been realized, yet there is a significant gap between ideal and realized efficiencies. For optimization, the current generation process must be analyzed. Due to the layered subcell design, it is impossible to physically contact only one subcell and measure its electrical properties, such as current–voltage curves. We proposed a new method to determine the



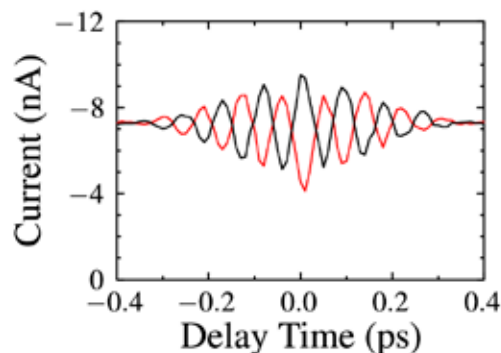
**Figure 2.** Power dependence of time-resolved photoluminescence decays from the middle subcell of a triple-junction solar cell.

subcell's electrical performance from the power dependence of time-resolved photoluminescence decays.

Since this method does not require any contact, all subcells can be probed individually. The charge separation and carrier recombination rates are determined from the photoluminescence decays, allowing us to understand the physics of the current generation in each subcell.

## Coherent Control of Photocurrent Dynamics in Bulk GaAs Using Phase-locked Pulses

Photocarrier generation in semiconductors is an essential process that determines the photon-to-current conversion efficiency. To analyze the carrier generation sites in semiconductors, we developed an ultrafast photocurrent spectroscopy using a phase-lock of excitation pulses. In this technique, the relative phase between two excitation pulses is locked to yield constructive or destructive interference. The ultrafast photocurrent dynamics are analyzed by measuring the photocurrent intensity as a function of the delay time between the excitation pulses. By analyzing the time-resolved photocurrent of bulk GaAs, we found that the shallow acceptors are dominant carrier generation sites at room temperature. Furthermore, the photocurrent intensity and direction were successfully controlled by changing the phase-lock conditions.



**Figure 3.** Photocurrent intensity under the constructive (black line) and destructive (red line) phase-lock conditions.